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DISPERSION OF CO_2 DROPLETS IN THE DEEP OCEAN

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ABSTRACT

Dispersion of CO_2 droplets in the deep ocean under the influence of currents and turbulence has been investigated. It is shown that: (1) the concentration of the CO_2 effluent can be described by Gaussian distributions under a distorted coordinate transformation; (2) ocean turbulence does not influence the direction of motion of the CO_2 droplets; however, it can exert a significant influence on effective droplet velocities; and (3) deep ocean currents alter the direction of droplet motion, distorting the droplet cloud. Both deep currents and marine turbulence can favorably affect CO_2 discharge in the ocean by enhancing dissolution and limiting local environmental impacts.

KEYWORDS

CO_2; contaminant; droplet; ocean; dispersion.

INTRODUCTION

Ocean disposal and sequestration of CO_2 captured from fossil fuel combustors is being evaluated as a means to slow its accumulation in the atmosphere. The majority of scenarios proposed to date call for discharge of pure, liquefied CO_2 at depths below the thermocline (i.e., below about 500 m). The behavior of the effluent following release in the deep ocean needs to be confirmed via experiment and modeling studies, since it will impact profoundly the extent of the hazard posed to the marine environment and the effectiveness of the concept as a greenhouse gas emissions countermeasure.

The CO_2/seawater system is hydrodynamically unstable; hence liquid CO_2 effluent will break up into a dispersed droplet phase (Masutani *et al.*, 1995; Nishikawa *et al.*, 1995). At ocean pressures greater than 44.5 bar and temperatures less than 283 K, a thin solid hydrate layer will rapidly encase the CO_2 droplets (Aya *et al.*, 1992; Teng *et al.*, 1995), impeding dissolution. Break up of the effluent and hydrate effects have been afforded inconsistent treatment in earlier studies of the fate of CO_2 discharged in the deep ocean (Golomb *et al.*, 1989; Golomb *et al.*, 1992; Liro *et al.*, 1992; Morishita *et al.*, 1993; Haugan *et al.*, 1995; Holder *et al.*, 1995; Kobayashi, 1995). The liquid effluent has been modeled both as a continuous phase (no break up) and a droplet plume; hydration of the CO_2 has been predicted to proceed to completion, been confined to a thin film at the seawater-CO_2 interface, or ignored.

In this study, it is assumed that liquid CO_2 discharged below 500 m will exist initially as a droplet cloud that can be assigned a "contaminant" concentration, C. The droplets comprising the contaminant phase are buoyant at depths above about 3000 m and dissolve slowly (due to the hydrate film) as they move through the water. While the behavior of dissolving CO_2 droplets has been modeled previously (see, for example, Golomb *et al.*, 1989; Liro *et al.*, 1992; Holder *et al.*, 1959; Haugan *et al.*, 1995), the effects of deep ocean currents and fluid turbulence have not been considered directly. If the droplets are less than 10 mm in diameter, then their buoyant velocities are of the same order of magnitude as the velocities typically associated with ocean eddies and deep currents (Robinson, 1983). This suggests that the dispersion of the droplets may be influenced significantly by ocean hydrodynamics.

To investigate the effects of ocean currents and turbulence on the transport of CO_2 released in the deep ocean, an analysis was conducted of the advection and dispersion of a contaminant droplet phase. The analysis considers a point (or line) release of liquid CO_2 that breaks up immediately to form a cloud of small, monodispersed droplets which do not agglomerate nor coalesce. To facilitate the analysis, the contaminant concentration is taken to represent all carbon initially derived from the effluent.

ADVECTION-DISPERSION WITH NEUTRAL BUOYANCY

To avoid buoyant rise of the CO_2 effluent, discharge at depths between 3000 and 3500 m has been proposed (Aya *et al.*, 1992; Ohsumi *et al.*, 1992; Lund *et al.*, 1994). At the temperatures and pressures which prevail at these depths, the difference in the densities of the liquid CO_2 contaminant and seawater are small; the CO_2 droplets may be considered neutrally buoyant and, therefore, amenable to classic dispersion analysis. Although this line of inquiry will not provide information on the motion of individual droplets, it can predict the spatial transport characteristics of the droplet ensemble.

In the deep ocean, stable vertical stratification leads to predominant horizontal (steady or unsteady) flows characterized by length scales much larger than those in the vertical direction. The following advection-dispersion equation describes the evolution of the contaminant CO_2 (or carbon) concentration:

$$\frac{\partial C}{\partial t} + u(z,t)\frac{\partial C}{\partial x} = k_x \frac{\partial^2 C}{\partial x^2} + k_z \frac{\partial^2 C}{\partial z^2} \quad . \tag{1}$$

Here, u is the (horizontal) ocean current velocity, and k_X and k_Z are, respectively, the horizontal and vertical dispersion coefficients. The point x = 0; z = 0 corresponds to the release site.

The ocean current velocity may be modeled as

$$u = u_0(t) + \alpha(t)z \ . \tag{2}$$

The linear shear distorts the droplet cloud. To account for this deformation, we introduce a distorted coordinate system, (X,Z,t), in which Z = z and

$$X = x - \int_0^t u_0(t')dt - zG_x(t) \ , \tag{3}$$

where G_X is a distortion parameter that must be determined. Under the new coordinates, (1) becomes

$$\frac{\partial C}{\partial t} + (\alpha - \frac{dG_X}{dt})z\frac{\partial C}{\partial X} = (k_x + k_z G_X^2)\frac{\partial^2 C}{\partial X^2} - 2G_X k_z \frac{\partial^2 C}{\partial X \partial z} + k_z \frac{\partial^2 C}{\partial z^2} \ . \tag{4}$$

To simplify the analysis, boundary conditions are assumed to be time-independent. If G_X is taken to be

$$G_x = \int_0^t \alpha(t')dt' \ , \tag{5}$$

then (1) takes the form of a time-dependent dispersion equation (Young *et al.*, 1982; Smith, 1982):

$$\frac{\partial C}{\partial t} = K_X \frac{\partial^2 C}{\partial X^2} \ . \tag{6}$$

The dispersion coefficient, K_X, is defined as $K_X \equiv k_X + k_Z G_X^2$. The solution of (6) is

$$C(X,t) = \frac{M}{\sqrt{2\pi\sigma_X^2}} \exp(-\frac{X^2}{2\sigma_X^2}) \ , \tag{7}$$

where M is the source strength of the discharge and σ_X^2 is a variance that satisfies $d\sigma_X^2/dt = 2K_X$. Under distorted coordinates, (7) indicates that dispersion of a CO_2 droplet cloud produces a Gaussian concentration distribution. The distortion parameter G_x depends on the ocean current: for steady currents, the shear factor α is constant and $G_x = \alpha t$ and $\sigma_X^2 = 2k_x t + (2/3)\alpha^2 k_z t^3$; for temporally periodic currents, $\alpha = \alpha'\cos(\omega t)$, leading to $G_x = (\alpha'/\omega)\sin(\omega t)$ and

$$\sigma_X^2 = 2k_x t + \frac{k_z}{2\omega}(\frac{\alpha'}{\omega})^2(2\omega t - \sin 2\omega t) \ ,$$

where α' and ω are the amplitude and frequency of the current, respectively.

ADVECTION-DISPERSION WITH POSITIVE BUOYANCY

Disposal system costs and limitations on submerged pipeline technology recommend discharge of liquid CO_2 at moderate depths between 500 and 1500 m (Herzog *et al.*, 1993). In this region, buoyancy of the contaminant must be considered. Under the constraints of the dispersion analysis, a vertical current is employed to account for buoyant transport. The functional form of this pseudo-current is inferred by considering the terminal rise velocity, V, of a CO_2 droplet, which is attained when fluid drag balances buoyancy:

$$g\Delta\rho\frac{4}{3}\pi r^3 = \frac{1}{2}C_D\pi r^2\rho_w V^2 \ . \tag{8}$$

In this expression, g is the gravitational acceleration, $\Delta\rho$ the density difference between seawater and CO_2, r the droplet radius , C_D the drag coefficient, and ρ_w the seawater density. An expression for the droplet velocity is derived from (8):

$$V = \left(\frac{8g\Delta\rho r}{3C_D\rho_w}\right)^{1/2} \ . \tag{9}$$

The drag coefficient may be estimated via the Stokes' relationship (White, 1991): $C_D = (12\mu_w)/(rV\rho_w)$. The droplet velocity then is given by $V = (2/9)(g\Delta\rho r^2/\mu_w)$, where μ_w is the seawater viscosity. In order to derive an expression for V as a function of location and time, consider the vertical displacement of the buoyant CO_2 droplets:

$$z = \int_0^t V dt' = \frac{2g\Delta\rho}{9\mu_w}\int_0^t r^2(t')\,dt' \ . \tag{10}$$

Here, it is assumed that the upper limit of integration, t, is less than the time required for complete dissolution of the droplets (at which point, buoyant rise is irrelevant). In addition, changes in fluid properties with depth are ignored. Since experiments suggest that the shrinkage rate of a CO_2 droplet in seawater, $A \equiv |dr/dt|$ is approximately constant (Aya *et al.*, 1992; Shindo *et al.*, 1995), (10) becomes

$$z = \frac{2g\Delta\rho}{27\mu_w A}(r_0^3 - r^3) \ , \tag{11}$$

leading to

$$r^2 = r_0^2\left(1 - \frac{27\mu_w A}{2g\Delta\rho r_0^3}z\right)^{2/3} \ ,$$

where r_0 is the initial radius of the droplets. It follows that

$$V = \frac{2g\Delta\rho r_0^2}{9\mu_w}\left(1 - \frac{27\mu_w A}{2g\Delta\rho r_0^3}z\right)^{2/3} \ . \tag{12}$$

Since A is very small (Aya *et al.*, 1992; Shindo *et al.*, 1995), (12) may be approximated as

$$V \approx V_0 + \gamma z \ , \tag{13}$$

where $V_0 = (2/9)(g\Delta\rho r_0)/\mu_w$ and $\gamma = -2A/r_0$.

As indicated by (13), the vertical velocity of the contaminant CO_2 phase depends only on position, decreasing linearly with increasing z. The evolution of the contaminant concentration, C, may, therefore, be described by an advection-dispersion equation

$$\frac{\partial C}{\partial t} + u(z,t)\frac{\partial C}{\partial x} + w(z)\frac{\partial C}{\partial z} = k_x\frac{\partial^2 C}{\partial x^2} + k_z\frac{\partial^2 C}{\partial z^2} \ , \tag{14}$$

where the velocity of the vertical pseudo-current, which accounts for buoyant transport, is modeled after (13) to be $w(z) \equiv w_0 + \beta z$. Once again, $u(z,t) = u_0(t) + \alpha(t)z$. The parameters α and β depend on the hydrodynamic conditions at the disposal site. Letting $C(x,z,t) = C_1(x,t)C_2(z,t)$ yields

$$\frac{\partial C_1}{\partial t} + u(z,t)\frac{\partial C_1}{\partial x} = k_x \frac{\partial^2 C_1}{\partial x^2} \tag{15}$$

and

$$\frac{\partial C_2}{\partial t} + w(z)\frac{\partial C_2}{\partial z} = k_x \frac{\partial^2 C_1}{\partial z^2} \ . \tag{16}$$

Applying a procedure similar to that used in the neutral buoyancy case, (15) and (16) are solved yielding

$$C_1 = \frac{M_X}{\sqrt{2\pi\sigma_X^2}} \exp(-\frac{X^2}{2\sigma_X^2}) \ , \tag{17}$$

and

$$C_2 = \frac{M_Z}{\sqrt{2\pi\sigma_Z^2}} \exp(-\frac{Z^2}{2\sigma_Z^2}) \ , \tag{18}$$

which leads to

$$C(X,Z,t) = \frac{M}{2\pi\sigma_X\sigma_Z} \exp\left[-(\frac{X^2}{2\sigma_X^2} + \frac{Z^2}{2\sigma_Z^2})\right] , \tag{19}$$

where $M = M_X M_Z$ is the source strength and X and Z are distorted coordinates given by

$$X = x - \int_0^t u_0(t')dt' - z\int_0^t \alpha(t')\,dt' \qquad (z \neq 0) \ ,$$

$$Z = z - \int_0^t w_0 dt' - z\left[(\beta z / w) + \exp(-wt / z)\right] \qquad (w \neq 0) \ ;$$

The variances are $\sigma_X^2 = 2K_x t$ (where, once again, $K_x \equiv k_x + k_z G_x^2$) and

$$\sigma_Z^2 = 2K_z \int_0^t (1 - G_Z^2)dt' \ ,$$

with $K_Z = k_z(1-G_z^2)$ and $G_z = \exp(-wt/z) + (\beta z/w)$. Equation (19) indicates that the dispersion of buoyant CO_2 droplets also produces a Gaussian distribution in the distorted coordinate frame. Noting that stable vertical stratification of the deep ocean produces $k_x >> k_z$, and considering the definitions of the variances, it is obvious that the effect of ocean turbulence on the dispersion of CO_2 effluent is manifested primarily through σ_X. As expected, strong turbulence (i.e., large values of k_x) results in large σ_X, enhanced spatial dispersion, and a small peak value of effluent concentration, C.

The concentration profile of the droplet cloud at x = 0, i.e., on the axis of the discharge, is of interest. At this locale, (19) becomes

$$C|_{x=0} = \frac{M}{2\pi\sigma_X^2\sigma_Z^2} \exp\left[-((\int_0^t u dt')^2 / 2\sigma_X^2 + Z^2 / 2\sigma_Z^2)\right] \ . \tag{20}$$

Tilting of the axis of the droplet cloud from vertical therefore is induced through a non-zero value of

$$(\int_0^t u dt')^2 / (2\sigma_X^2) \ ;$$

i.e., by the ocean current. For a steady ocean current,

$$\left(\int_0^t u\,dt'\right)^2 / (2\sigma_X^2) = \frac{u^2 t}{4k_x} \;; \tag{21}$$

for a periodic ocean current,

$$\left(\int_0^t u\,dt'\right)^2 / (2\sigma_X^2) = \left(\int_0^t u_0(t')\,dt' + \frac{\alpha'}{\omega} z \sin\omega t\right)^2 / 4k_x t \;. \tag{22}$$

DISCUSSION AND CONCLUSIONS

While the preceding analysis is not relevant to individual CO_2 droplets, it demonstrates clearly that currents and ocean turbulence can exercise significant influence on the motion of the droplet ensemble. If buoyancy can be neglected, as, for example, in some of the very deep ocean disposal scenarios, then from (7), the concentration of the effluent assumes a one-dimensional Gaussian distribution under distorted coordinates. Transport effects are manifested primarily through the variance term. For a steady ocean current, the variance can be expressed as $\sigma_X^2 = 2k_x t + (2/3)\alpha^2 k_z t^3$. Since the growth of σ_X is nonlinear with respect to time, the dispersion of the droplets is not purely horizontal. Shearing by the ocean current may induce a tilting of the axis of the droplet cloud. In general, for a temporally periodic current, the variance also grows nonlinearly with time; however, at large values of t, $2\omega t >> \sin 2\omega t$, and, thus, $\sigma_X^2 = 2K_x t$, where $K_x = k_x + (k_x/2)(\alpha'/\omega)^2$. The parameter α'/ω can be quite large at some locations (Gargett *et al.*, 1981), resulting in the effective dispersion coefficient K_x exceeding k_x by a significant amount.

The CO_2 effluent concentration also assumes a (two-dimensional) Gaussian distribution under distorted coordinates when droplet buoyancy is non-negligible. Examination of (19) and (20) indicates that turbulence effects, which are manifested through the dispersion coefficients, k_x and k_z, combine with currents to alter the effective droplet velocities and reduce peak values of C (through the variances). Turbulence, however, does not deflect the droplet cloud; the tilting of the cloud axis arises solely through the action of currents.

The conclusions of the present study may be summarized as follows: (1) analytical solutions can be obtained for the concentration of a CO_2 contaminant being dispersed and advected by deep ocean turbulence and currents through the application of a distorted coordinate transformation; the contaminant concentration is described by one- (neutral buoyancy) or two-dimensional (buoyant) Gaussian distributions in the distorted coordinate frame; (2) turbulence does not influence the direction of the droplet motion but may alter effective droplet velocities during dispersion; (3) currents induce changes in the direction of the droplet motion, tilting the axis of the droplet cloud; and (4) these induced changes in the effective velocities and direction of the droplet cloud can enhance mass transfer and limit local impacts on the marine environment; hence, ocean turbulence and currents may favorably influence marine disposal of CO_2.

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